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Plutonium Residue Recovery (PuRR) Project Quarterly Progress Report (Initial Report through March 1988)

**L. C. Pittenger, R. M. Alire, M. S. Coops,
R. E. Priest, D. S. Thompson, R. H. Condit,
L. Burris, and R. D. Pierce**

June 27, 1988



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Foreword

The Plutonium Residue Recovery (PuRR) project addresses the recovery of weapons-grade plutonium from intractable residues that are currently stored at several Department of Energy sites.

This project is a joint effort by Lawrence Livermore National Laboratory (LLNL) and Argonne National Laboratory, and has two primary objectives:

- Efficient plutonium recovery from feedstocks that currently cannot be processed.
- Rejection of tramp elements in a waste form that contains < 100 nCi/g of Pu (< 1.6 ppm Pu), i.e., non-transuranic ("non-TrU") waste.

More than 2 metric tonnes of Pu have accumulated in intractable residues since the 1950s. The amount of intractable Pu residue has averaged about 50 to 100 kg per year. The cost-effective technology in LLNL's PuRR proposal is now deemed feasible for cleaning up heretofore intractable Pu residues. The results will be a lessening of potential hazards to the environment, cleanup of vault overloading with no need for enlarging national vault capacity, and the return of usable Pu to the pipeline.

We gratefully acknowledge the contributions of J. A. Leary (a Los Alamos National Laboratory retiree) of TRU Engineering Company, a consulting firm, who helped define the PuRR process, and of D. W. Gregg, Chemistry and Materials Science Department, LLNL, who helped work out the details of the process steps. We also thank Donna Lee Paquette for editing the report and preparing the manuscript.

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Plutonium Residue Recovery (PuRR) Project Quarterly Progress Report (Initial Report through March 1988)

Abstract

The PuRR project integrates a number of well understood pyrochemical concepts into one functional process for the recovery of plutonium from scrap and residues. Critical process steps have been identified. Confirming experiments are now in progress at Lawrence Livermore National Laboratory (LLNL) and Argonne National Laboratory (ANL) to demonstrate the successful application of the pyrochemical process described in this document.

I. Introduction

The Plutonium Residue Recovery (PuRR) project is a research and development effort to design and implement a process for the recovery of plutonium from scrap and residues resulting from the production of weapons-grade material. Its objectives are: (1) the efficient recovery of plutonium of high enough purity for recycle to the production lines; and, (2) the reduction of the plutonium content of the residues to a level (< 100 nCi/g) at which they may be disposed of, economically, as non-transuranic ("non-TrU") waste.

The ongoing production of metallic plutonium products has resulted in an accumulation of scrap and residue materials containing a large quantity (estimated at several tonnes) of weapons-grade plutonium. Although the plutonium concentration may vary widely from sample to sample, the average concentration of plutonium is approximately 3% by weight. Thus, these residues not only constitute a valuable store of plutonium, but comprise a large volume of hazardous waste that requires expensive storage facilities, and for which the cost of high-level disposal would be ruinous. Current processing of these residues amounts to highgrading, i.e., much of the residue has been subjected to traditional acid leaching treatment. Thus, what remains is indeed "intractable" as regards conventional processing; and, while significant plutonium has been extracted, the amount remaining is sufficient to war-

rant the development of a process to recover it with the objective of finally reducing the residue to non-TrU activity.

The PuRR project has been entered into jointly with ANL. Much of the technology of pyrochemical processing was developed at ANL in the 1950s and 1960s in connection with reprocessing of fuel from the Experimental Breeder Reactor No. 2 (EBR-II). Pyrochemical and electrochemical processes are currently being developed at ANL for processing spent metallic fuel from the Integral Fast Reactor (IFR). ANL will provide basic research in support of this program, as well as demonstrate some of the unit operations on a laboratory scale, to provide proof-of-principle.

LLNL will conduct development of processes on engineering and production scales, with the ultimate goal of establishing and operating a pilot module capable of processing 100–200 kg of Pu per annum, prior to the transfer of technology for construction and operation of a production facility at one or more of the already established processing sites. This laboratory has had considerable experience in the pyrochemistry of plutonium; it is utilized as the balance-of-plant technology by the Special Isotope Separation (SIS) program being developed here. An additional, invaluable asset is the class I Plutonium Facility, Bldg. 332, already in place, and with space available for an effort of this magnitude.

II. Summary

This has been a period of developing and refining concepts of the PuRR process to utilize pyrochemical techniques for the recovery of plutonium from intractable residues. Major activities during this period are summarized below; more complete descriptions are given in the topical sections.

It became apparent early on that the relatively low (approximately 3%) concentration of plutonium in the feedstock, along with large quantities of other metallic species, would make early gross separation of the plutonium very desirable in order to lessen the amounts of molten salt and metallic solvents required by the process. The process as earlier conceived has been expanded to incorporate molten salt transport (MST) to move the plutonium selectively from a donor alloy [the solvent for direct oxide reduction (DOR)] to an acceptor alloy [the anode for solvent electrorefining (SER)]. The present PuRR process has been worked out in considerable detail, as will be described below.

Experiments performed at ANL indicate that the formation, during DOR, of intermetallics between plutonium and the aluminum and silicon comprising the bulk of the residues will not be a serious impediment.

Conditions for the reactivation of gloveboxes in the Bldg. 332 Plutonium Facility have been established. These boxes will be used to perform laboratory-scale demonstration-of-feasibility and proof-of-principle experiments. The conditions are incorporated in a set of action items, of which half have already been carried out.

Design requirements for the laboratory-scale studies' equipment have been set, and detailed design work is proceeding. Layouts have been produced for the design of new boxes for supporting engineering-scale experiments.

The development of first-cut process flow sheets has begun, for the case where MST is the mechanism for gross separation, as well as for alternate procedures involving chlorination of Pu, or Pu, Al and Si from the oxides.

Conceptual design work on a pilot plant has been shelved in favor of addressing the immediate need for check-out and development of experimental equipment by establishing a (radioactively) cold facility in which all glovebox conditions can be duplicated.

A survey of the literature to explore alternatives to the mainline recovery scheme has been carried out. Also, thermodynamic data pertaining to the mainline process has been gathered.

III. Process Development

During this period, techniques for handling intractable residues have been identified and organized into the proposed PuRR process. PuRR is designed to accept intractable residues of widely varying composition containing weapons-grade plutonium, currently stored at several Department of Energy sites, and process it with two primary goals: (1) the efficient recovery of plutonium; and (2) the rejection of tramp elements in a waste form containing < 100 nCi/g (< 1.6 ppm) Pu, i.e., non-TrU waste.

The above objectives are of primary importance in defining the PuRR process. Additionally, it is desirable that the process be one that: can be carried out with minimal radiation exposure for operators, will be compatible with computer-based process control options and current inventory interrogation requirements, and have high reliability and be simple to maintain. The latter implies incorporation of modular process equipment.

The major steps in the proposed process are shown in Fig. 1. All feed materials are first sub-

jected to ignition in air at 1200°C . This calcining of the wide variety of residues that will comprise the feed produces an organic- and carbon-free product consisting primarily of oxides, oxyhalides, and oxysulfates, which will be compatible with the next process stage. This mixed oxide is then pulverized and subjected to gamma spectroscopy for determination of fissile content.

The next major step is the reduction of the mixed oxides to the metallic state, and dissolution of the mixed metals into a liquid metal alloy solvent.

Next, a gross separation of a concentrated actinide fraction from the bulk of other elements is effected by a selective high temperature liquid-liquid extraction (pyroextraction) process. An MST process has been chosen for this stage.

Final product recovery is accomplished by SER from a liquid metal alloy solvent anode to produce pure liquid plutonium.

The process outlined has been selected to achieve the goals stated earlier; in particular, it is

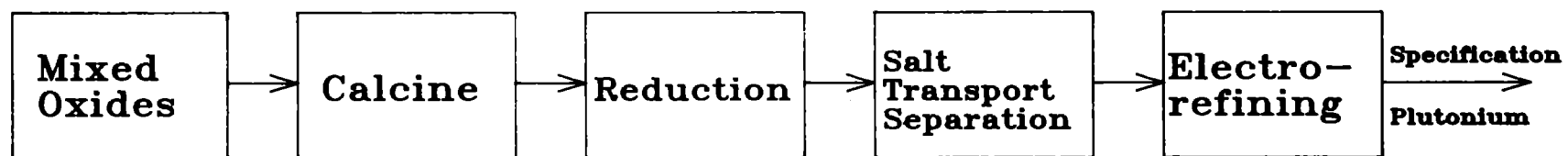


Figure 1. Main steps of the PuRR pyro-extraction process.

anticipated that >75% of the waste generated will be non-TrU.

The process is shown in somewhat more detail in Fig. 2. The individual process steps shown are soundly based.¹⁻³ Each operation is backed by well-established thermodynamics. Several of the operations have already been performed on a production basis, and most of the others have been demonstrated in the laboratory. There remain, however, two technical issues to be resolved. These are: (1) the effect of large amounts of silicon and aluminum on the salt/donor alloy distribution coefficients; and (2) the extent to which it will be possible to strip the waste reagents (salts used as solvents in the reduction stage and in the molten salt transport stage, and loaded donor and acceptor alloy constituents) of actinide alpha-emitting contaminants.

The reduction technique shown was chosen specifically to handle the PuRR feedstock, low (<5%) in plutonium, and with a wide variety of other metallic species. It is common practice to utilize DOR of plutonium oxide (PuO_2) to the metal as the first step in processing, with collection of the plutonium in the bottom of the crucible in liquid form at a temperature slightly above its melting point. This process will not result in a fluid product in the presence of gross impurities. Therefore, a solvent alloy is used to dissolve all the metallic products of reduction at approximately 800°C. Copper-60 wt% magnesium is chosen as the solvent, as it is suitable as a donor alloy in the subsequent MST stage. In common with DOR practice, calcium is the reductant, its oxide being soluble in the CaCl_2 flux to approximately 18 mol %. The presence of CaF_2 has been found to promote phase disengagement.

The reduction stage is shown in more detail in Fig. 3. It is estimated that the three counterflow stages will suffice to assure complete reduction of all the PuO_2 , and decontaminate the salt waste sufficiently to allow disposal as non-TrU waste.

The next stage in the PuRR process is the gross separation of actinides from tramp metals by MST. This step allows the final purification of plutonium by SER to be carried out with a greatly reduced quantity of anode alloy than would be the case if the electrolyrefining were to follow the reduction directly.

The basis for the molten salt transport stage is shown in Fig. 4. Distribution constants for plutonium and uranium between Cu-Mg and Zn-Mg al-

loys and a MgCl_2 -30 mol % NaCl -20 mol % KCl transport salt are shown plotted against Mg content of the alloys. Two facts are salient: the ratio between the distributions for the donor (Cu) and acceptor (Zn) alloys is three orders of magnitude over a wide range of Mg content. Also, the distribution coefficient between the donor and the salt, which is of order unity for actinides (and lanthanides), is much lower for the tramp metals (not shown in the figure).

The salt transport process works as follows: the Cu-Mg alloy, containing all the metallic products of the reduction step, is contacted with the transport salt. Lanthanide and actinide elements are transferred to the salt with distribution coefficient of order unity, but tramp elements remain behind. The loaded salt is then contacted with the Zn-Mg acceptor alloy. Plutonium and uranium will be extracted to a very large degree (with distribution coefficients of order 10^{-3}) into the acceptor. The bulk of the americium, which has a somewhat larger distribution coefficient between salt and acceptor than do plutonium and uranium, will remain in the salt.

The MST stage is shown in greater detail in Fig. 5. The donor alloy is first stripped of residual Ca from the reduction stage. After it has been contacted several times (only one step is shown) with the transport salt, a final stripping with CuCl_2 will reduce it to a non-TrU contamination level. The stripping reaction is thermodynamically driven by a net free energy change of -94.74 kcal/mole of Pu and -99.78 kcal/mole of Am.

The transport salt is reusable for many cycles before it becomes excessively burdened with americium and rare earths. It is then stripped by contacting with Ca-Mg alloy. The reaction is favored by a free energy change of -55.26 kcal/mole of PuCl_3 .

The physical moving of the alloys and salts can be accomplished by any of several means. Perhaps the simplest is pressure/vacuum transfer through a heated tube. This is particularly suited for laboratory development, and may also be used in a production-scale plant. The scheme is illustrated in Fig. 6. The tube is resistively heated by application of low voltage between its ends. (Both vessels are electrically isolated to permit this.) The adaptor at the receiver end ensures that current flows through the tube's entire length. It has been observed in the laboratory that the difference in conductance between an alloy- and

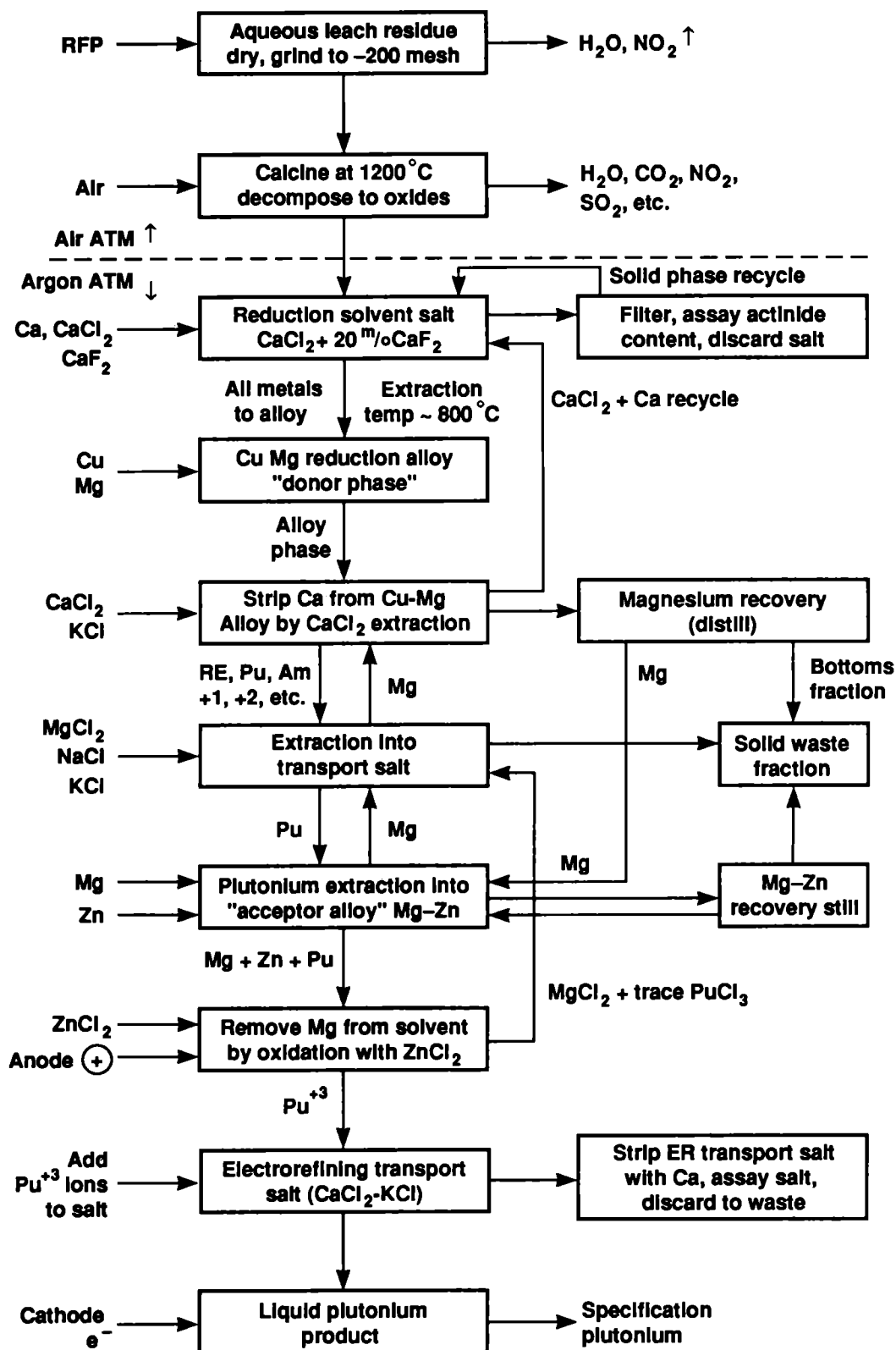


Figure 2. PuRR chemical schematic.

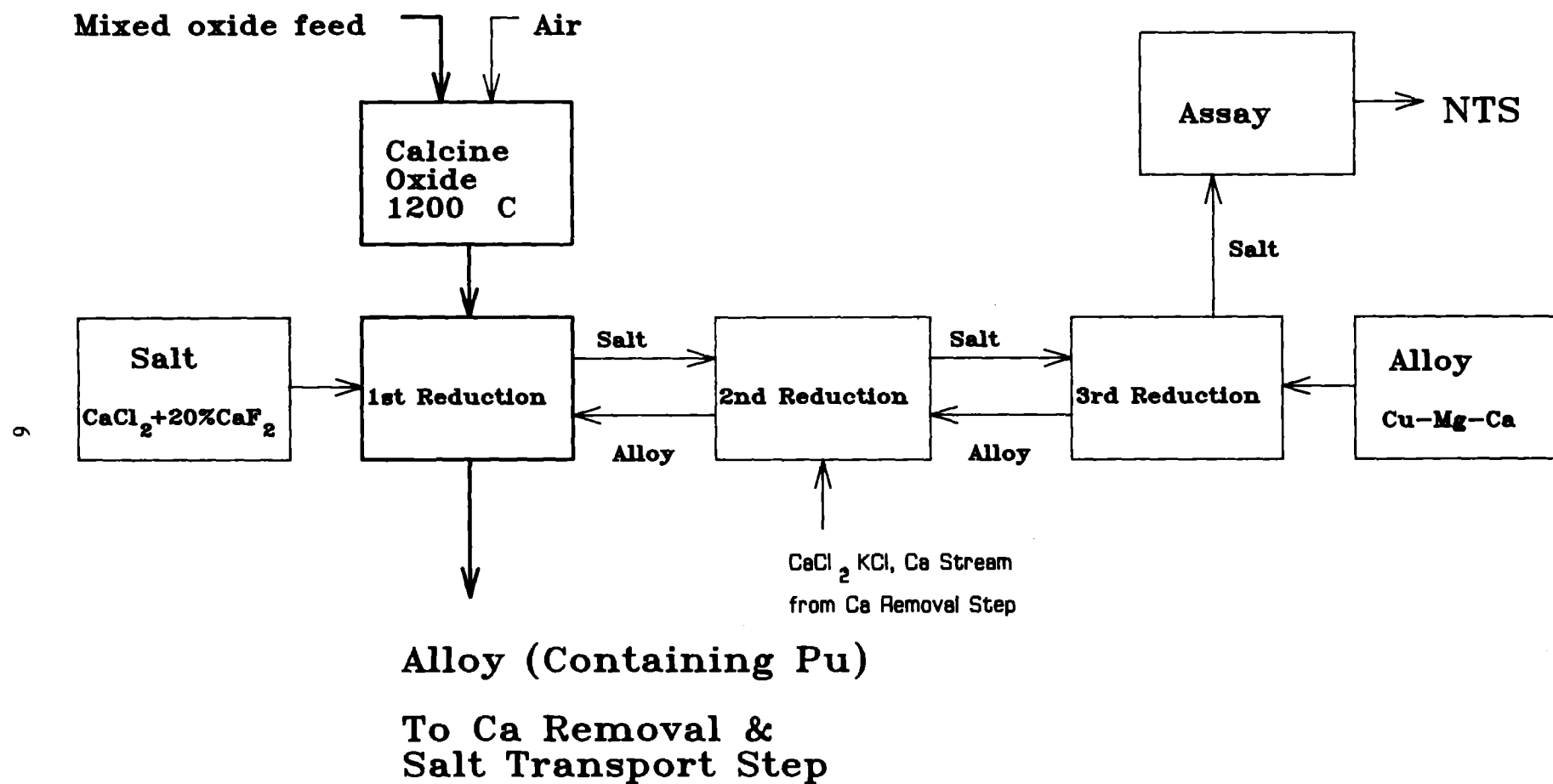


Figure 3. Calcining and reduction stages.

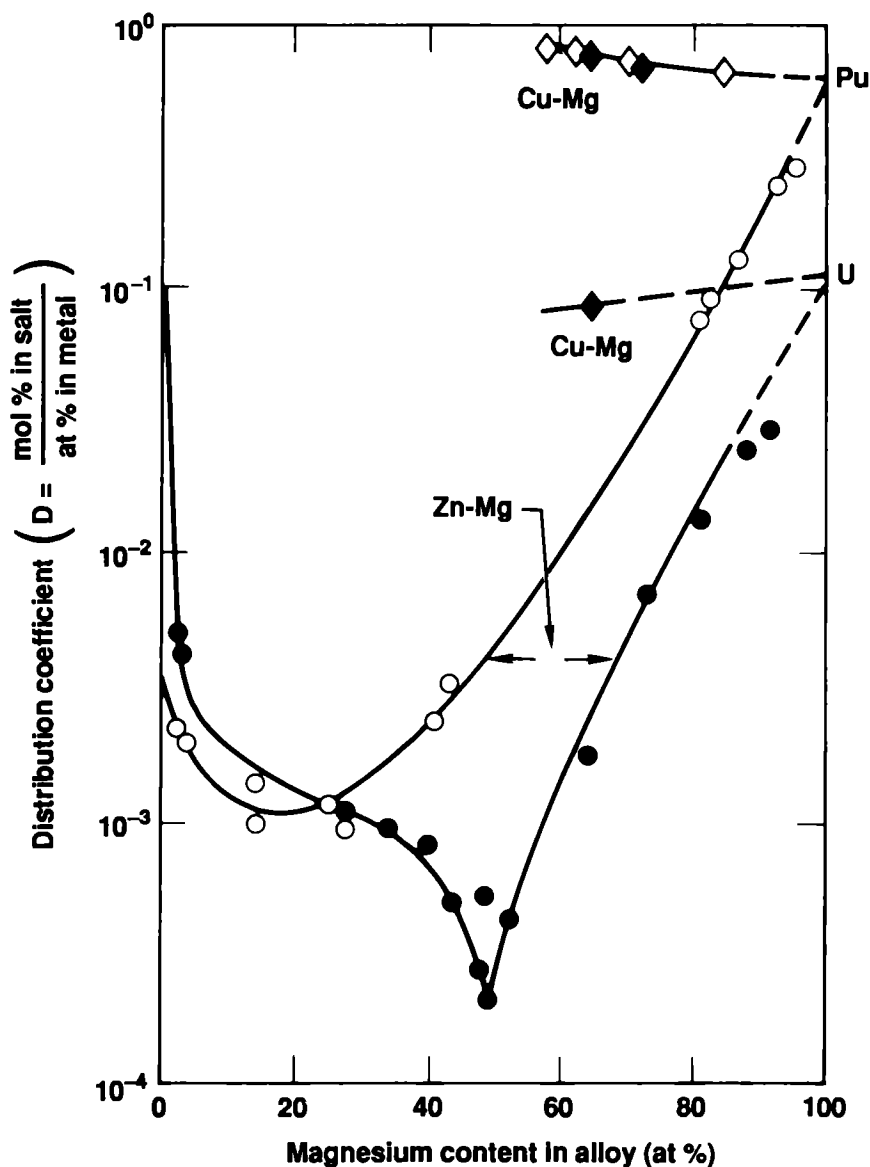


Figure 4. Distribution of U and Pu between 50 mol % MgCl_2 -30 mol % NaCl -20 mol % KCl salt and Cu-Mg, CD-MG, and Zn-Mg alloys at 600°C. (From J. B. Knighton, I. Johnson, and R. K. Steunenberg, "Uranium and Plutonium Purification by the Salt Transport Method," in *Symposium on Processing of Nuclear Fuels, Nuclear Metallurgy*, Vol. 15, CONF-690801, USAEC, Oak Ridge, TN, 1969.)

salt-filled length of tube changes the heating rate sufficiently to provide visual indication of the position of the interface, thus allowing for very complete transfer of one component without unwanted carryover of the other. The detection of the interface could be automated for use of this technique in a production plant.

A possible means of effecting the MST procedure using pressure/vacuum transfers is shown in Fig. 7. Note that, with each vessel electrically isolated from the others, only the tube being used will be heated. The unheated tubes are frozen closed and act as isolation valves.

The stripped donor alloy and transport salts are filtered and analyzed for alpha content prior to disposal.

The last major step in the PuRR process is the solvent electrorefining of the plutonium. This stage is shown in more detail in Fig. 8. The acceptor alloy from the MST stage is first stripped of Mg. The resulting alloy of zinc and plutonium with trace impurities is the anode of the electrorefining cell. (It is to be noted that refining out of a solvent permits the removal of essentially all of the plutonium without the complication of leaving behind a fraction in the form of a high-impurity

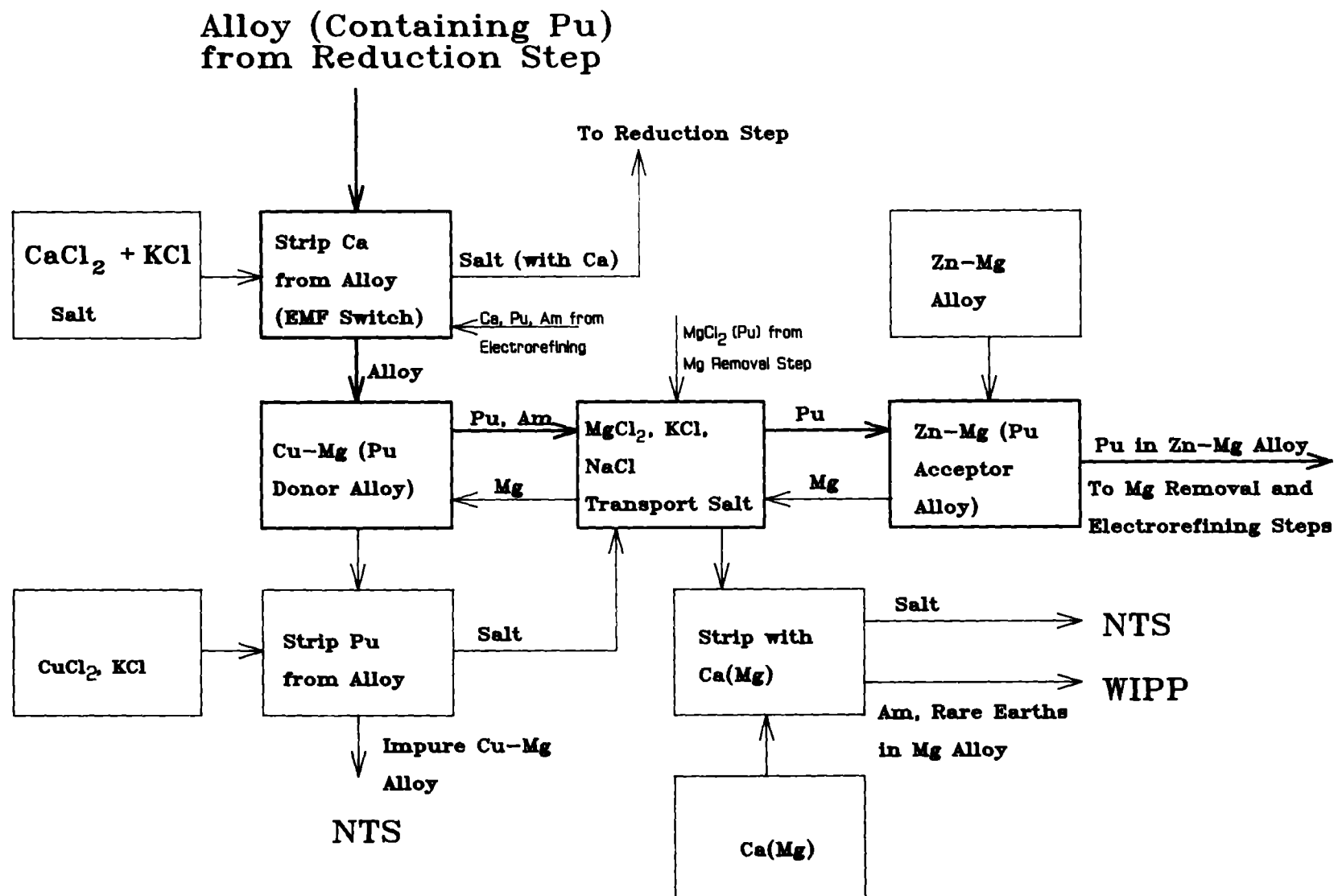


Figure 5. Ca removal and MST.

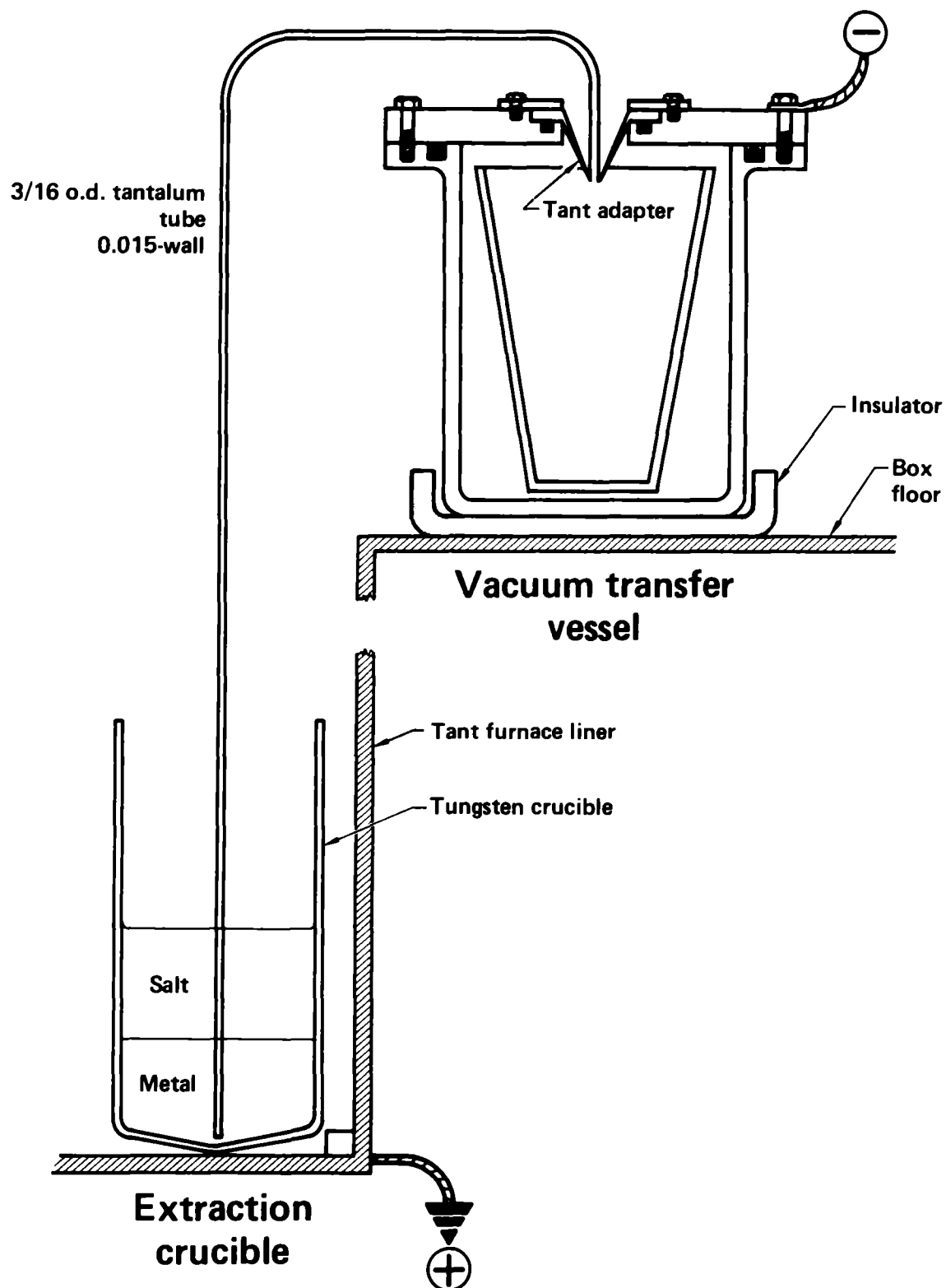


Figure 6. Typical arrangement for transfer of molten reagents.

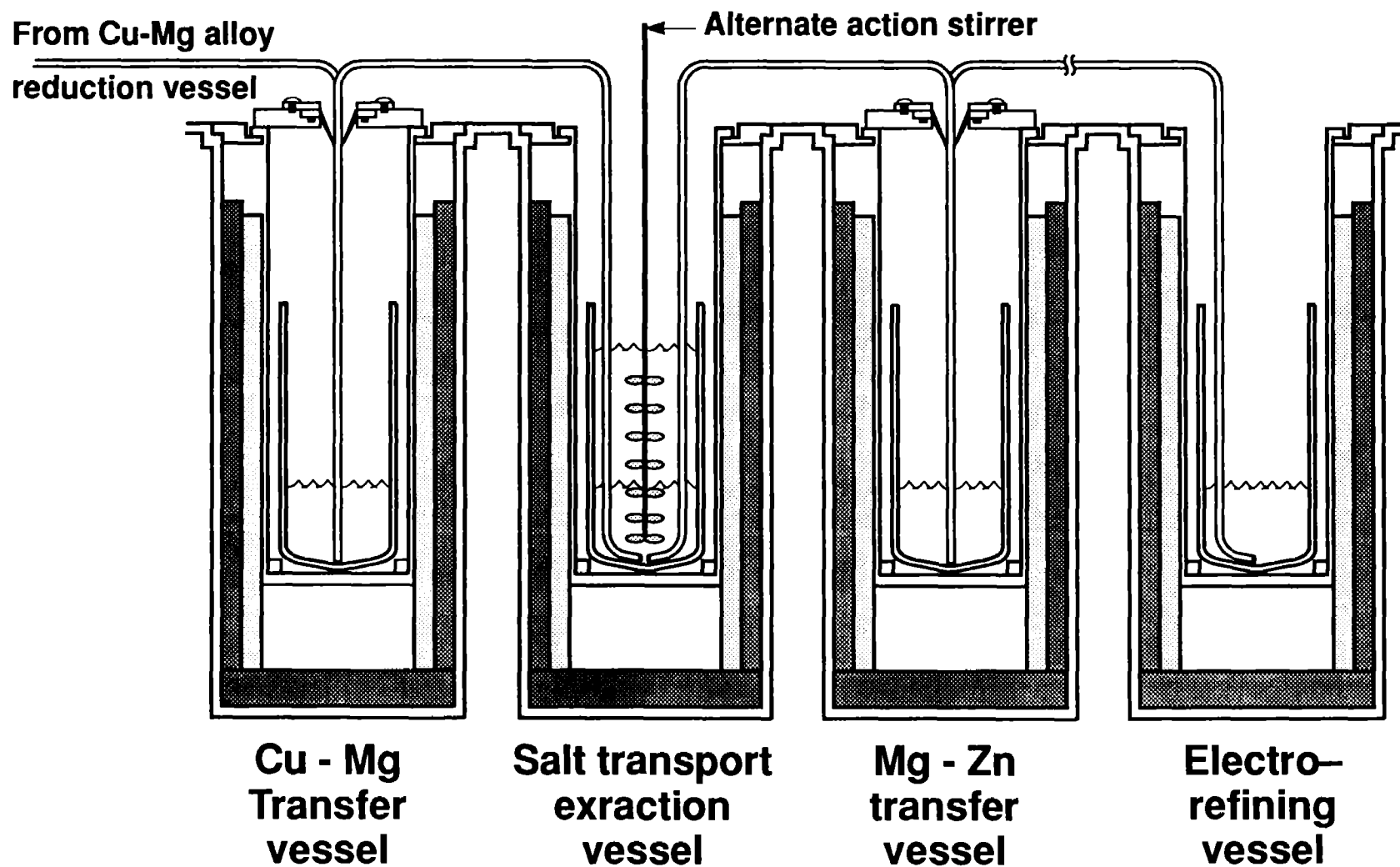


Figure 7. Molten alloy or molten salt transfer between any two tungsten reaction vessels by pressure or vacuum transfer line. All vessels are electrically isolated from each other.

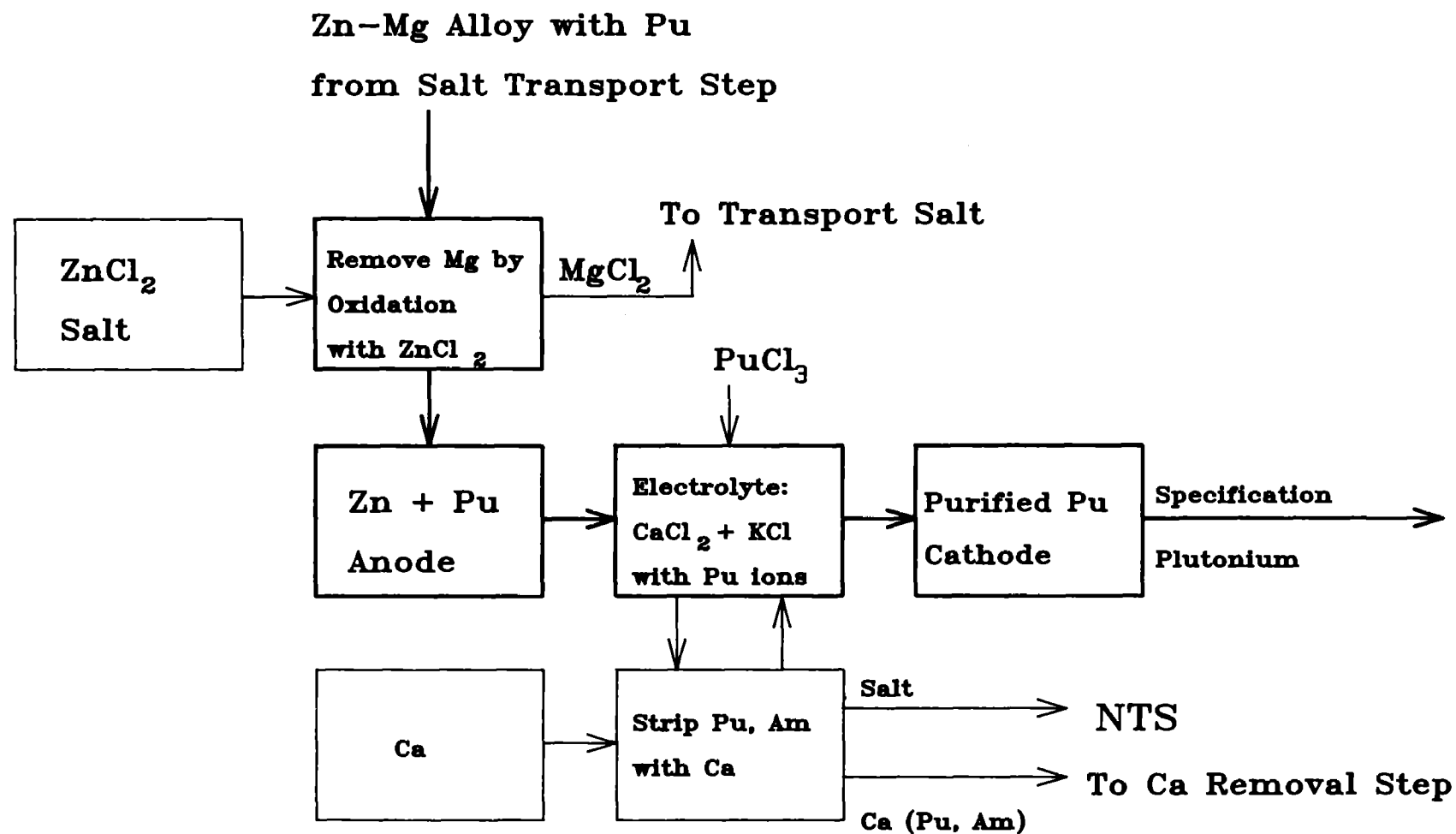


Figure 8. Mg removal and solvent electrorefining.

heel.) The electrolyte consists of a thermodynamically stable molten salt of $\text{CaCl}_2\text{-KCl}$ to which PuCl_3 is added. Pure liquid plutonium collects at the cathode.

The chart, Fig. 9, shows the anode and cathode half-cell reactions as well as a listing indicating typical disposition of impurities in electrorefining. It is obvious that a very pure plutonium product is attainable.

The solvent electrorefining process will require a somewhat different arrangement than is typical for refining from a slightly impure plutonium anode. Figure 10 shows the schematic of equipment proposed for semi-continuous SER. The solvent anode feed is transferred into the cell via heated transfer lines (as earlier described for the MST process). The purified product is removed similarly. Such a system can be operated nearly continuously, with batch sizes of 4-5 kg Pu. A layout of such a cell might be as shown in Fig. 11 in which reservoirs for feeding of anode and electrolyte and receiving of plutonium are shown solid, with reservoirs for accepting depleted anode and electrolyte (eventually loaded with Am) shown dashed.

Figure 12 shows the PuRR process in its entirety, with the disposition of all products and by-products indicated. Of significance is that only the americium need be disposed of as TrU waste for this (idealized) process.

In the next quarter, we shall continue to refine the definition of the PuRR process. Solubilities of species in the presence of other species will be examined to aid in establishing required quan-

Anode: $\text{Pu (impure)} \rightarrow \text{Pu}^{+3} + 3\text{e}^-$				
Cathode: $\text{Pu}^{+3} + 3\text{e}^- \rightarrow \text{Pu (pure)}$				
Net reaction: $\text{Pu (impure)} \rightarrow \text{Pu (pure)}$				
Behavior of Impurity Elements in Electrorefining				
Element	$-\Delta F^\circ$, kcal/gram atom Cl	Concentration, ppm ^b		Element concentrated in
		Feed	Product	
Ni	18	750	<10	Anode
Cu	21	100	2	Anode
Ta	22	5,000	80	Anode
Fe	27	25,000	20	Anode
Cr	32	280	<10	Anode
Ga	32	10,000	<25	Anode
Mn	41	70	<2	Anode
Al	46	2,700	<5	Anode
U	54	275	<20	Anode
Th	59	160	<10	Anode
Pu	59	—	—	—
Ce	66	527	<25	Salt
Am	67	850	82	Salt

Figure 9. Electrorefining chemistry.

ties of reagents and developing material balances and flow sheets. The relative uncertainties of various aspects of the process will be evaluated to guide the experimental work at ANL and that which will be done here in answering the most important unknowns.

IV. Laboratory-Scale Demonstration of Feasibility

Initial reduction experiments performed on residues at ANL have been inconclusive regarding completeness of reduction because of experimental difficulties. Indications are, however, that the reduction from residues is essentially complete. Particularly significant has been the demonstration of complete reduction of PuO_2 from a fired LECO crucible, as these crucibles are probably the most intractable constituents of the intractable residues we will handle.

The possible formation of intermetallics between Pu and Al, and Pu and Si has been of concern, as it was thought that the silicide in particular might render the Pu inaccessible to processing

downstream of the DOR stage. An experiment at ANL has shown that this need not be a concern. It was found that:

1. The addition of Al to a solution of Pu in Zn-10% Mg in an Al/Pu molar ratio of over 200 had no effect on the amount of dissolved Pu.

2. The addition of Si to a similar solution in a Si/Pu ratio of 54 *did* reduce the dissolved Pu by about 60%, **but: the introduction of Al into this mixture resulted in the complete re-dissolution of the Pu.**

Thus, although silicon has the potential for tying up some of the plutonium, this action will be inhibited by the presence of aluminum, which is

Side view

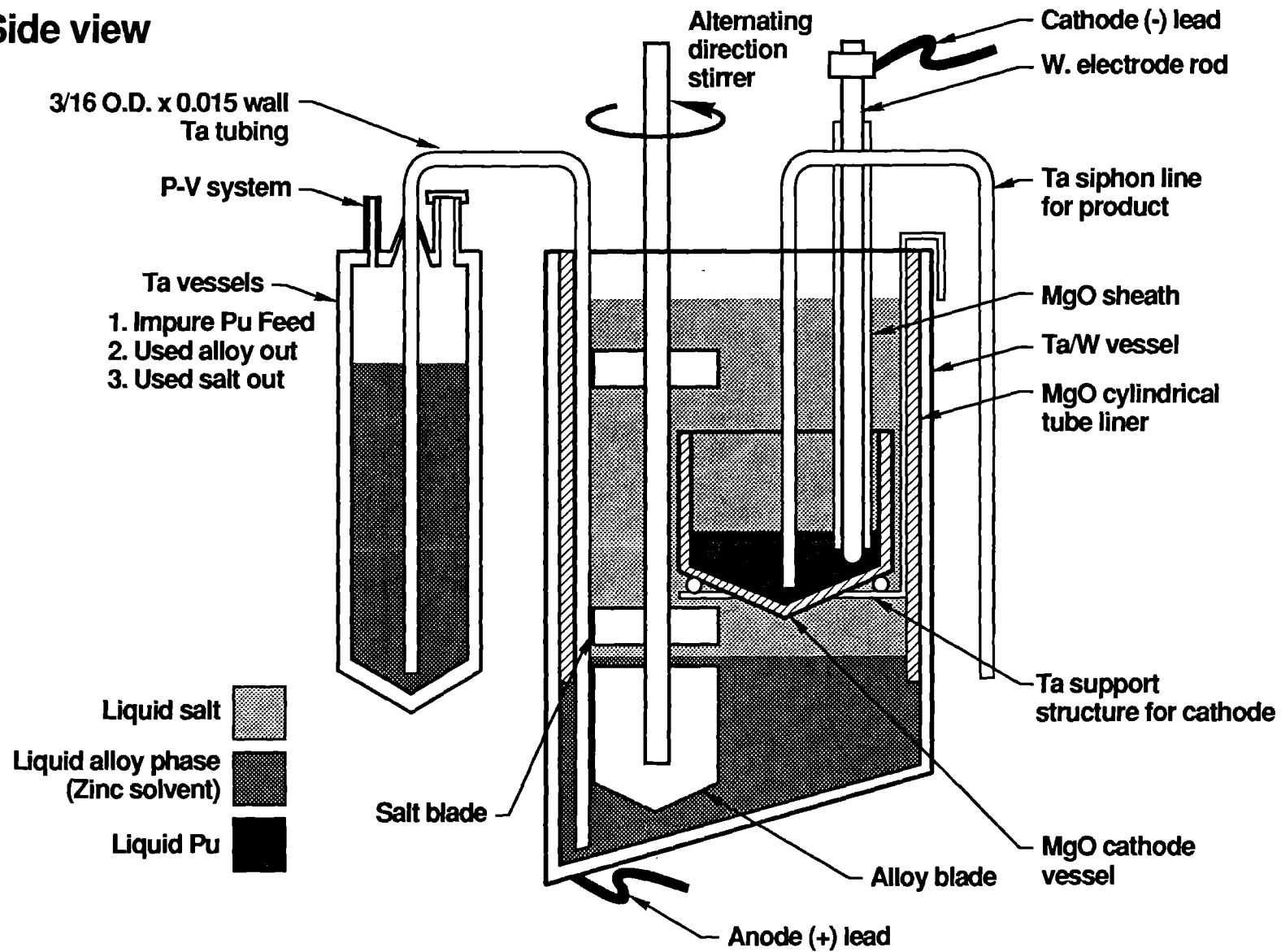


Figure 10. Semi-continuous SER system (cutaway).

Plan view

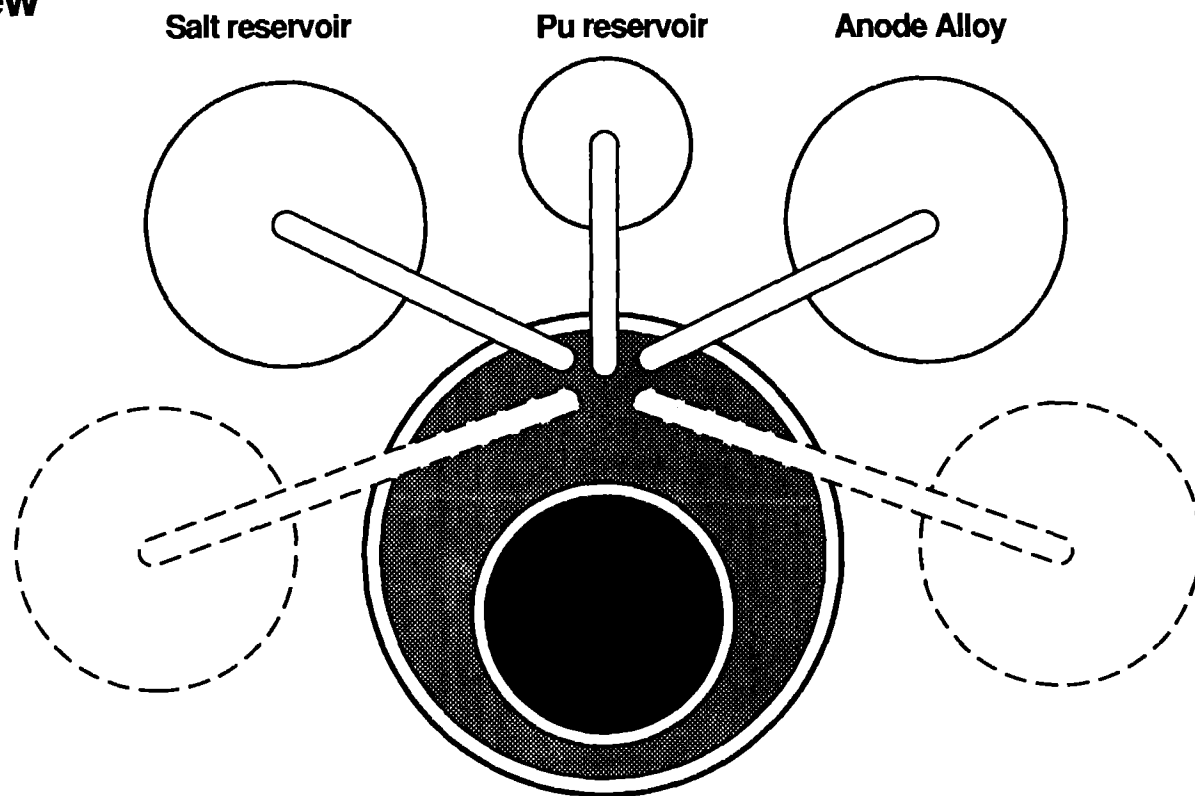


Figure 11. Semi-continuous SER system (layout).

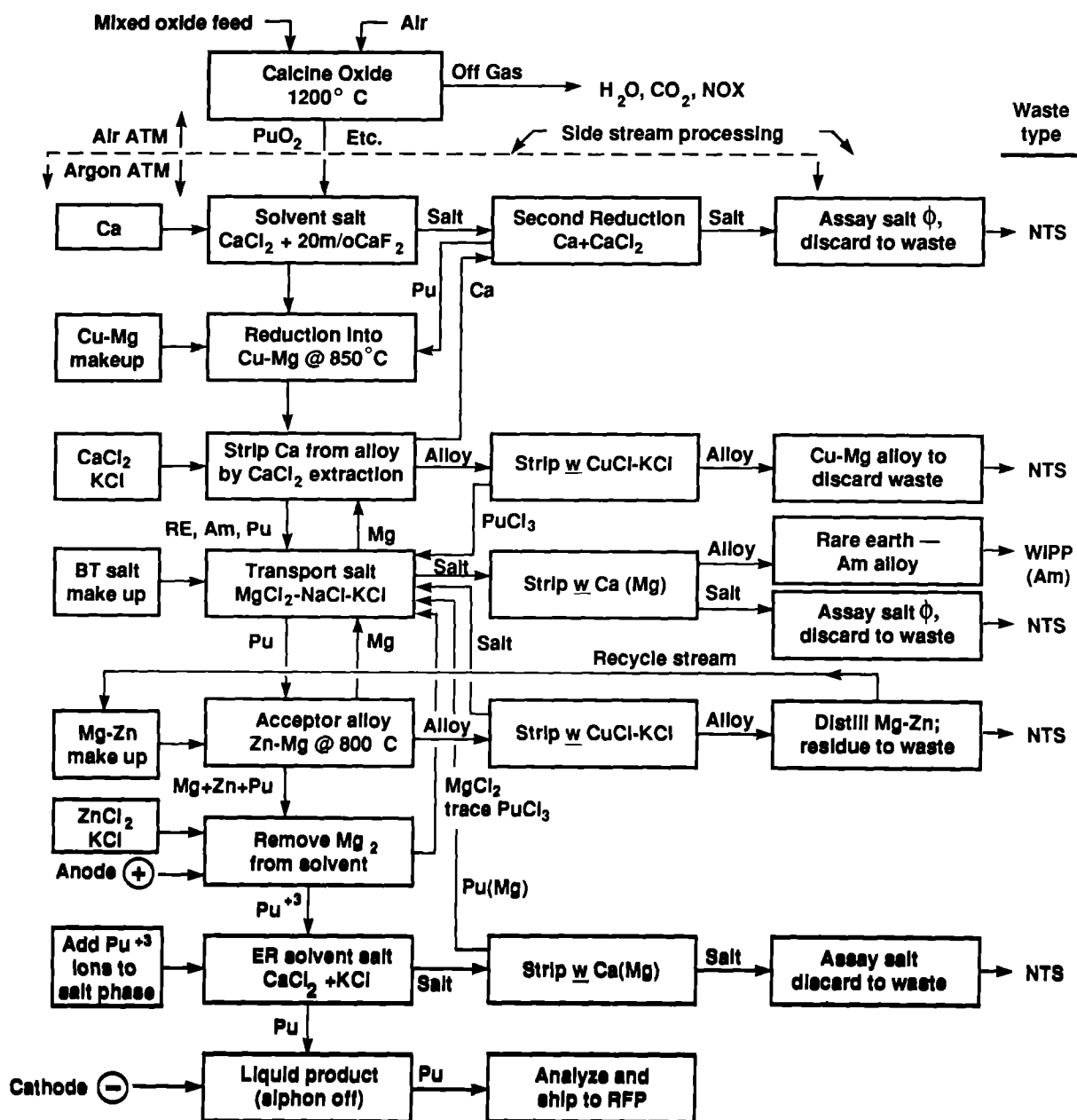


Figure 12. The PuRR process.

expected to be a major constituent of the residues as well.

In the next reporting period, we expect to demonstrate quantitatively the reduction of pluto-

nium from mixed oxide feed prepared from actual residues, and to attempt to determine the effect of aluminum and silicon on the distribution coefficients upon which the MST process depends.

V. Laboratory Demonstration of Engineering Support

Initial experiments at LLNL will be performed in existing pyrochemistry gloveboxes in Bldg. 332. Agreement has been reached with the

facility manager regarding action items to be completed prior to the reactivation of this equipment. The pyroline will require the reissuance of

its Operational Safety Procedure (OSP), upgrading of specific mechanical hardware, and outlining procedures for operation of the inert atmosphere purifiers.

One workbox will require the initial issue of an OSP, addressing seismic safety issues, and writing a Mechanical Engineering Safety Note to cover hydraulic press operations. This box will be equipped with a pulverizer and calcining furnace. A differential pressure gage is to be installed across its exhaust HEPA filter.

Boxes for salt preparation and aqueous operations require hard plumbing to the building ventilation system. The salt box requires replacement of a photohelic gage. The aqueous box needs a differential pressure gage, and the box's associated hoist bracket must be tested.

Approximately one-half of these requirements have been satisfied in this reporting period.

Memoranda were issued covering design requirements for equipment to support the initial PuRR experiments and initial experiments on the processing of Site Return Recycles (SRR). Equipment design is proceeding.

Initial design layouts for larger, engineering-scale experiments have been done. A conceptual furnace design for this scale of operations has been developed. Cost/availability comparisons are being done to determine which of various refractory metals and/or ceramics should be considered for solvent containment in the larger-scale experiments. In the next reporting period, we expect to complete all action items required to activate the existing gloveboxes. Experimental equipment designs will be finalized, subjected to review, and procurements initiated. Design for larger-scale experimental equipment will be advanced beyond the conceptual stage.

VI. Process Engineering

Preliminary process flow sheets are being developed for the mainline process that involves MST, to reduce the quantities of solvent required downstream of the DOR stage. Flow sheets for alternative methods of gross separation of Pu involving selective or non-selective chlorination (with separation by volatility in the latter case) are also being developed.

Size and cost estimates for production plant equipment are being developed concurrently. Present uncertainty as to the feedstock composition, the solubilities of the possible reduction products in the various proposed solvent mixtures, and the final selection of procedures for the production plant make close estimates of size and

cost impractical at present. It is apparent, however, that the amount of salt required to suspend the oxides in the initial DOR (or chlorination) stage will be large, and trade-offs between batch sizes and parallel processing stages are indicated.

Activity in the next reporting period will depend upon the degree to which the main process steps can be decided upon, based upon laboratory-scale experiments at LLNL and ANL, as well as the degree of confidence with which the solubility of various species in processing salt and metal solvents can be predicted. It is expected that progress in these areas will permit the development of more detailed process flow sheets as well as equipment size and cost estimates.

VII. Pilot Plant and Cold Check-Out Facility

Early in this reporting period, it became apparent that pilot plant studies could and should be delayed pending firmer definition of the production process, and that effort expended toward the expedition of laboratory-scale proof-of-principle experiments would better utilize the limited project resources. Accordingly, the remainder of this period has been spent in establishing a cold-test facility. This facility will permit the testing of newly acquired equipment under inert atmosphere glovebox conditions, at process temperatures, and with process reagents, before committing it to installation in the hot boxes in

Bldg. 332. It is anticipated that this facility will eventually become a place for the development of production-scale processes, the testing of procedures and equipment, and the training of pilot plant operators.

A location in the Heavy Element Facility, Bldg. 251, has been found, and formally applied for through the facility manager. All major items of equipment required for the initial check-out operations have been located in surplus storage, and have been acquired on one year or longer loans at no cost to the project. Designs of modifications to the surplus glovebox to accommodate

the initial experimental equipment have begun. In the next period, we expect to have refurbished the surplus gas purifying units, and to have com-

pleted the design of glovebox modifications and begun fabrication.

VIII. Study of Alternate Procedures

The necessity for further study of thermodynamic properties of various binary oxides and mixed oxide solutions has been established by reviewing published reports on the Synroc program at LLNL as well as standard reference works on the actinides.

We have reviewed the literature concerning methods of chlorination of silicon and aluminum, since these relatively volatile chlorides could be easily separated from the nonvolatile trichloride of plutonium. None of the chlorination methods studied is completely without drawbacks, but some development would most likely result in an acceptable procedure, should this approach prove necessary or desirable.

As a departure from pyrochemistry, we have also looked at the possibility of using synthesized

actinide-specific chelating agents for converting plutonium into a soluble form. Most of the current research into this is at a very low funding level and is aimed primarily at health physics applications. It appears that the compounds being studied, while adequately stable for use in very controlled medical applications, may not be stable enough for industrial chemistry environments.

In the next period, we expect to expand our compilation of data on Pu phase diagrams, in order to more closely estimate the quantities of salt and metal solvents that will be required in a production plant. This is in support of the process engineering activities reported on earlier. We also expect to carry out a systematization of estimates for thermodynamic properties of binary oxides and oxide solutions.

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